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THE HEALTH PHYSICS ASPECTS OF URANIUM PROCESSING

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ABSTRACT

The general problems associated with the health physics aspects of uranium processing are described. The physical characteristics of the radiations from uranium are presented along with a summary of the biological aspects of uranium exposure. Typical analytical equipment and techniques are described as well as the nuclear instrumentation required to provide the necessary area and personnel monitoring. A discussion of established methods of exposure control is also included.

THE HEALTH PHYSICS ASPECTS OF URANIUM PROCESSING

Introduction

The industrial processing of uranium is somewhat complicated by the fact that uranium is both a radioactively and chemically toxic material. As such, special techniques are required in the handling of this material in order that the associated hazards may be adequately controlled. Years of experience in both AEC research laboratories and production facilities have shown that these techniques are not overly restrictive and that entirely adequate control can be achieved by the application of good initial design coupled with sensible operation. The intent of this paper is to describe in more or less general terms the considerations that have evolved through the years and which have resulted in a program which strikes, without mutual compromise, an equitable balance between economy and safety.

Physical and Biological Characteristics

The radioactive decay of each of the important isotopes of uranium is characterized by the emission of an alpha particle. This process, which leads to the formation of an isotope of the element thorium, is shown in Figure 1. As can be seen, the alpha decay is followed by one or more beta decays each of which is accompanied by another change of species. The ultimate end of this series of transformations is in each case a stable isotope of lead, but since this process requires millions of years, it is of academic interest only. In general, it is the emission of the alpha particles which makes

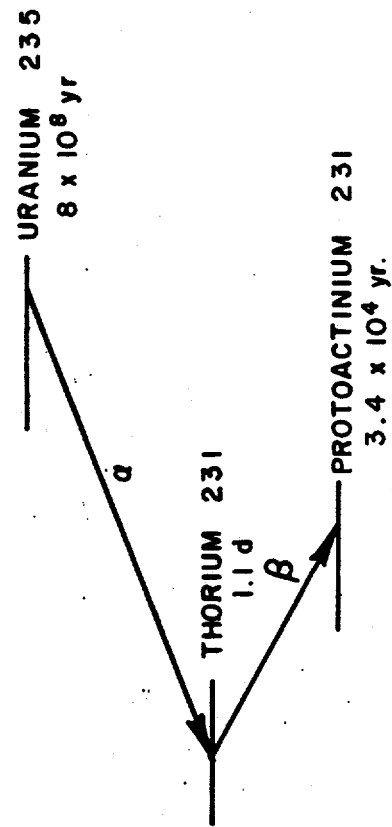
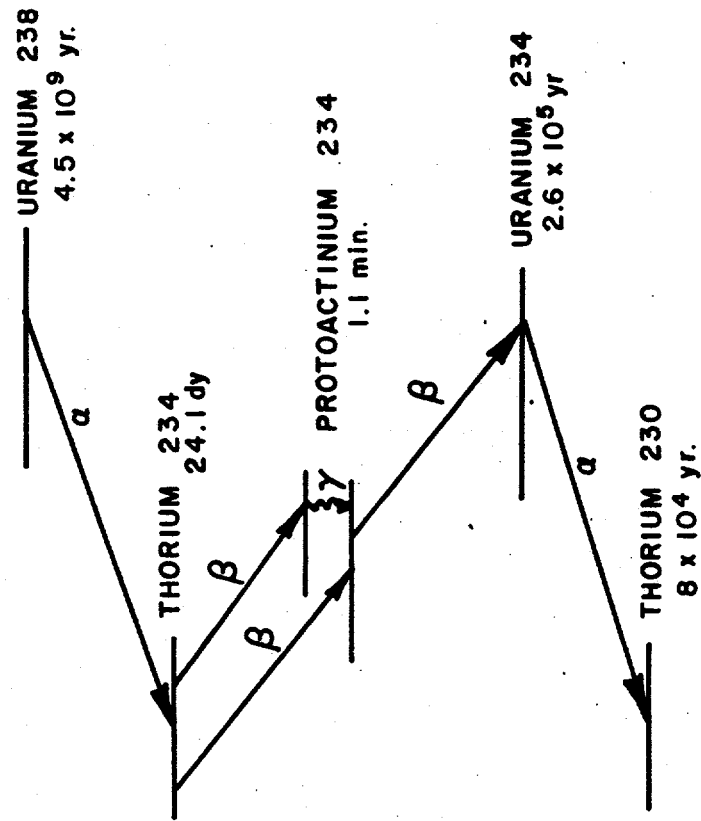
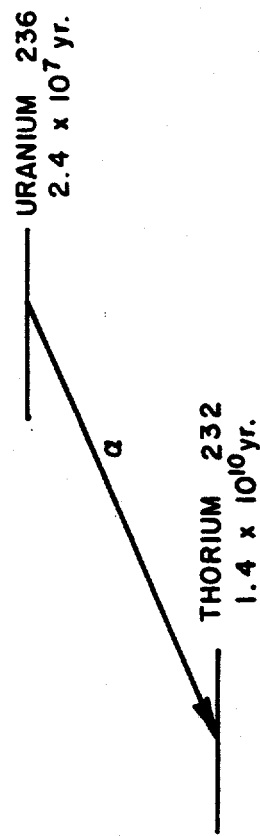


FIGURE 1. DECAY SCHEMES OF THE URANIUM ISOTOPES

uranium a radiological hazard. The associated beta disintegrations and their accompanying gamma rays, because of their much higher penetrating power, present a lesser degree of hazard when contained within the body but a significantly greater hazard when external to the body. Since the range of the uranium alpha particles is only about 50 microns in tissue, the associated radiation hazard is negligible until the material is taken into the body. However, once inside, the hazard is considerably magnified because of the dense ionization associated with the alpha particles.

Since the specific activity, or the activity per unit mass, varies inversely as the half life of a radioisotope, each isotope of uranium has a different degree of radioactivity. The method of determining the specific activity is shown in Figure 2 along with the calculated values for the uranium isotopes of interest. Figure 3 shows the activity relationship between these isotopes and can be used to determine the specific activity for any composite isotopic mixture provided the composition of the mixture is known. The total specific activity is merely the sum of the specific activities of the components as read on the ordinate of the graphs. It can easily be seen that the isotope uranium 234 is the controlling factor in almost any composition, in fact, it accounts for about 49 per cent of the total activity in normal isotopic uranium and over 95 per cent in typically enriched uranium.

The chemical toxicity of uranium depends on the solubility of the compound; the more soluble being more toxic. As with most heavy metals, the primary

ISOTOPE	HALF LIFE (yrs.)	SPECIFIC ACTIVITY	
		$\mu\text{C}/\mu\text{gm}$	$\text{d}/\text{m} - \mu\text{gm}$
URANIUM 234	2.475×10^5	6.180×10^{-3}	13,720
URANIUM 235	7.13×10^8	2.14×10^{-6}	4.74
URANIUM 236	2.391×10^7	6.387×10^{-5}	141.8
URANIUM 238	4.507×10^9	3.338×10^{-7}	0.741

$$\text{SPECIFIC ACTIVITY} = 1/m \times dN/dt = \lambda N/m$$

WHERE λ = DECAY CONSTANT = $\log_e 2$ / HALF LIFE

N = NUMBER OF ATOMS = AVOGADRO'S NUMBER / ATOMIC WEIGHT \times MASS

$$\text{CURIE} = 3.700 \times 10^{10} \text{ DISINTEGRATIONS PER SECOND}$$

FIGURE 2. RADIOACTIVE DECAY CHARACTERISTICS OF THE URANIUM ISOTOPES

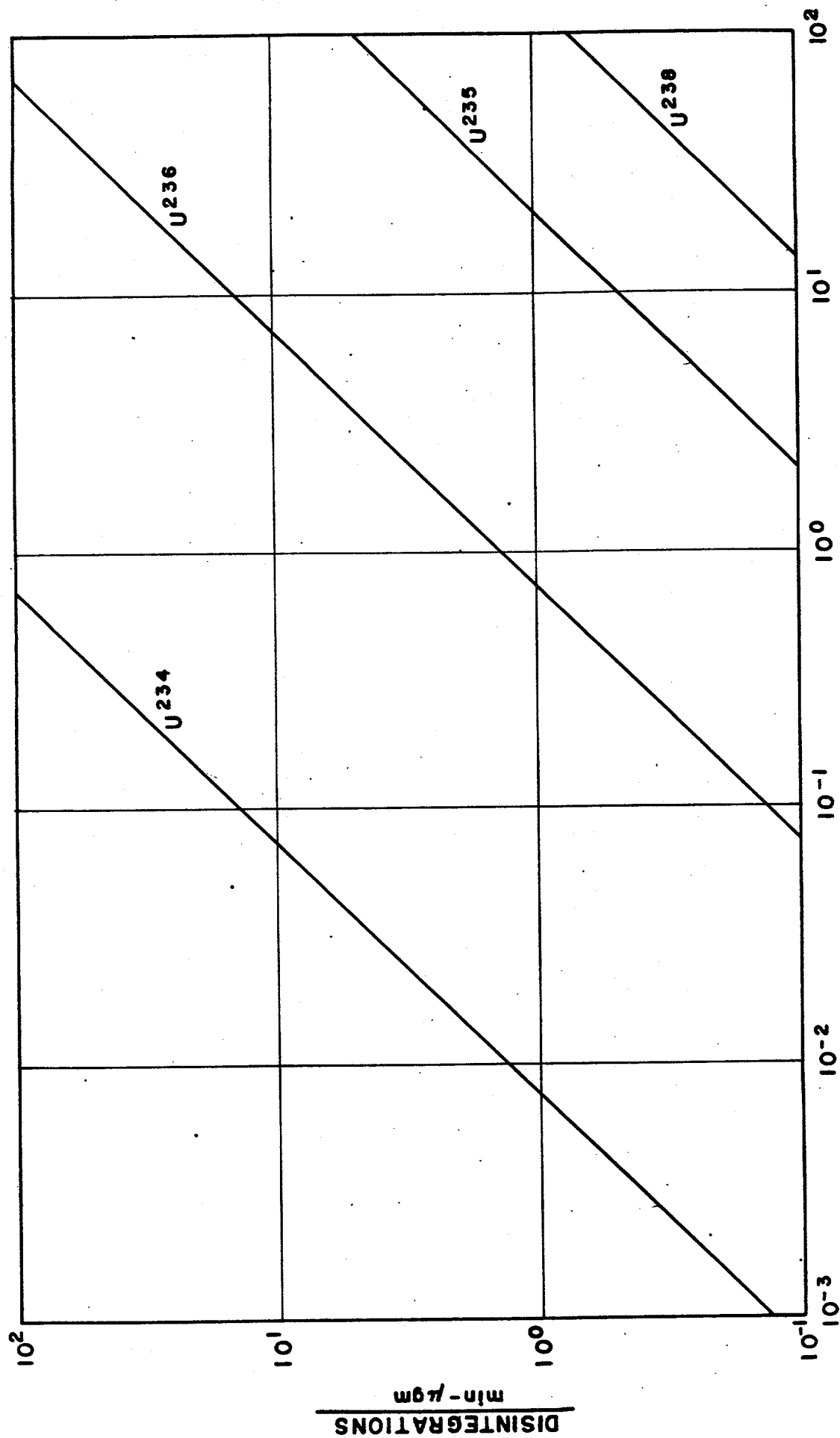


FIGURE 3. RELATIVE ACTIVITIES OF THE URANIUM ISOTOPES

effect is on the kidney. Even though uranium is perhaps one of the more toxic of all poisons, relatively large amounts can be tolerated since it is absorbed by the body to only a small degree. The radioactivity of uranium further complicates matters because now the biological effect is not necessarily mass dependent but, as we have seen, depends primarily on the uranium 234 concentration. Here the solubility, route of entry and, to some extent, the valence state determine the internal distribution.

Figure 4 shows the distribution of various forms of uranium in the body.

The radiobiological effects of internally deposited uranium are associated with the effects on a so-called critical organ which is defined as that organ receiving radiation damage which results in the greatest over-all damage to the body. It is generally the organ which contains the highest concentration of the uranium. As has been shown, for soluble normal isotopic uranium, the kidney becomes the critical organ. For insoluble material, both normal and enriched, the lung becomes the critical organ while for soluble enriched material the bone is the critical organ. The radiation effects on these parts of the body may, in general, be expected to be similar to the well published effects associated with other types of radiation exposure.

The maximum permissible exposure limits for radioactive materials are generally based on the recommendations of two recognized groups; the National Committee on Radiation Protection and the International Committee on Radiation Protection, functioning under the auspices of the National Bureau of Standards and the International Congress of Radiology, respectively.

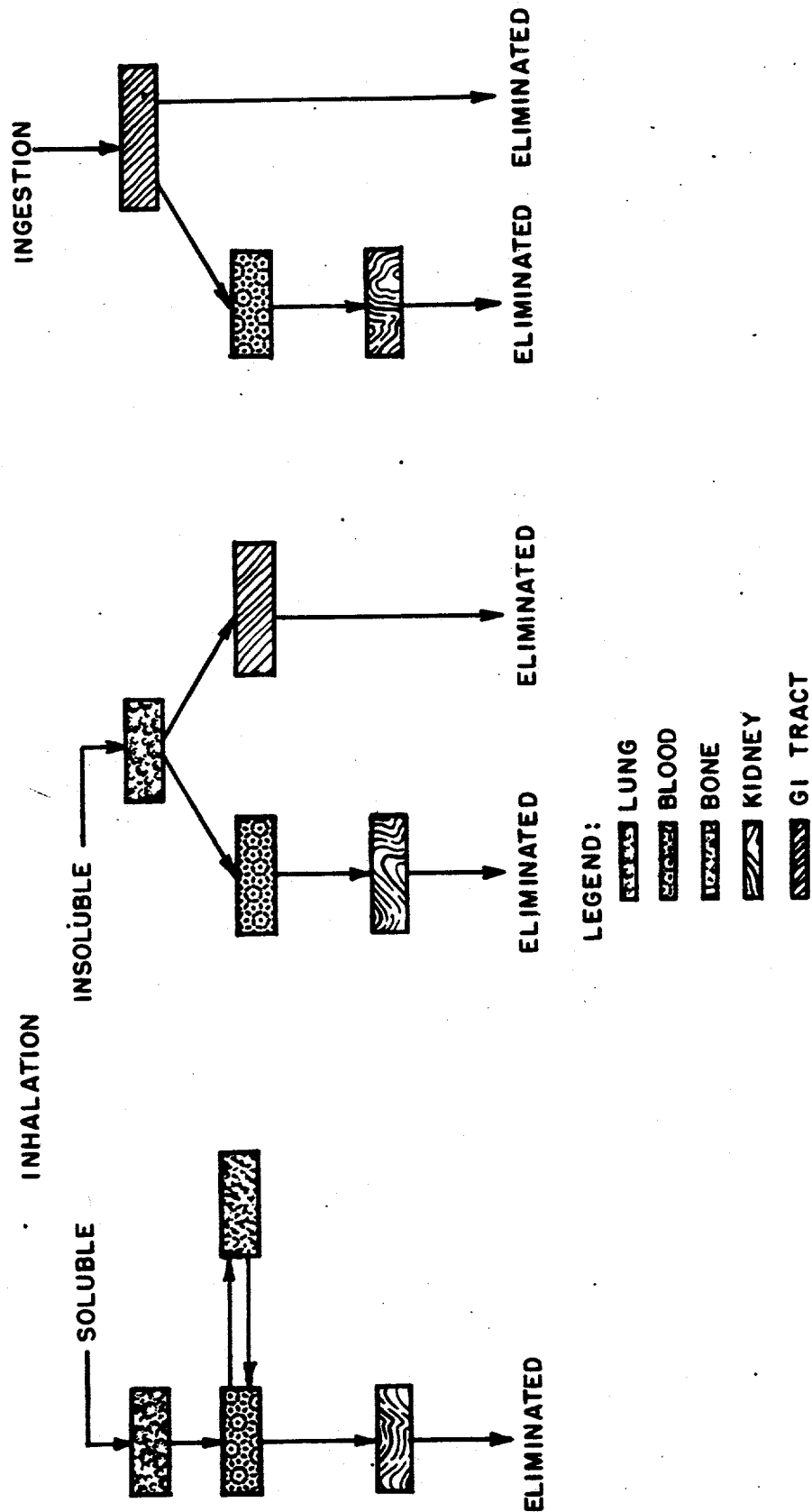


FIGURE 4. DISTRIBUTION OF URANIUM WITHIN THE BODY

The basic limits for radiation exposure are founded, in part, on the many unfortunate experiences which occurred during the early history of X-ray and radium usage. In some cases, the establishment of these levels is aided by animal exposure data, but this is generally limited to studies of distribution and excretion of internally deposited materials. With uranium, the situation is on firmer ground since some human exposure cases are available to support the values obtained from animal data extrapolation. Once the distribution pattern is known, it becomes possible to calculate the excretion rate corresponding to the permissible body burden. In making the calculations, generally a simple exponential elimination is assumed. This oversimplification is used for ease of application since elaborate calculating equipment would be required to handle a large number of exposures treated by a more exact excretion model. The maximum permissible limits recommended by the National Committee on Radiation Protection for air and water along with the excretion limits calculated by the method described by Neuman⁽¹⁾ are shown in Figure 5.

Methods of Analysis

Analyses of the atmospheric contamination in the operating areas are generally used to assess the degree of hazard resulting from the inhalation of uranium or its compounds. There are many ways in which this can be done, but perhaps the most widely used is the filtration method in which standard paper filters are used to remove the particulate matter. The

TYPE OF MATERIAL	MEDIUM		
	AIR	WATER	URINE
NORMAL ISOTOPIC SOLUBLE URANIUM	$5.1 \times 10^{-11} \mu\text{C/ml}$	$2.1 \times 10^{-4} \mu\text{C/ml}$	$14 \mu\text{g / day}$
NORMAL ISOTOPIC INSOLUBLE URANIUM	$5.1 \times 10^{-11} \mu\text{C/ml}$		$25 \mu\text{g / day}$
TYPICALLY ENRICHED SOLUBLE URANIUM	$3.4 \times 10^{-11} \mu\text{C/ml}$	$1.7 \times 10^{-3} \mu\text{C/ml}$	$8.9 \times 10^{-5} \mu\text{C/day}$
TYPICALLY ENRICHED INSOLUBLE URANIUM	$5.1 \times 10^{-11} \mu\text{C/ml}$		$1.5 \times 10^{-5} \mu\text{C/day}$

FIGURE 5. MAXIMUM PERMISSIBLE LIMITS FOR OCCUPATIONAL EXPOSURE

collected material is then analyzed by measuring the number of alpha particles produced by the radioactive decay of the uranium. If the efficiency of the filter and counter is known and the volume of air passing through the filter has been measured, then it becomes a relatively simple matter to calculate the magnitude of the air-borne contamination. Figure 6 shows three typical samplers utilizing filter collection systems while an exploded view of one type of filter holder is shown in Figure 7. Another method of collecting particulate matter utilizes the electrostatic precipitation principle. Here the particles are electrically charged in passing through a corona discharge and then collected on the surface of a metal cylinder or disk by electrostatic attraction. The material is then radioassayed in much the same manner as before. Both of these methods are sensitive down to about 1/10 of the maximum permissible limit, but they suffer from one common disadvantage. Since all particulate matter is collected, the radioactive daughter products of radon and thoron are deposited along with the uranium. These materials, which arise from minute amounts of radioactive contaminants in the ground, are always present in an undistributed atmosphere. By a gross radiometric analysis, these materials are also counted and because of their very high specific activity can, at times, completely mask the uranium activity. Fortunately, because of their short half life, a waiting period of 24 hours allows for almost complete decay but also prevents the final analysis of the uranium contamination prior to the end of the 24 hour period. Alpha pulse analysis will circumvent the delay, but this requires electronic equipment not generally

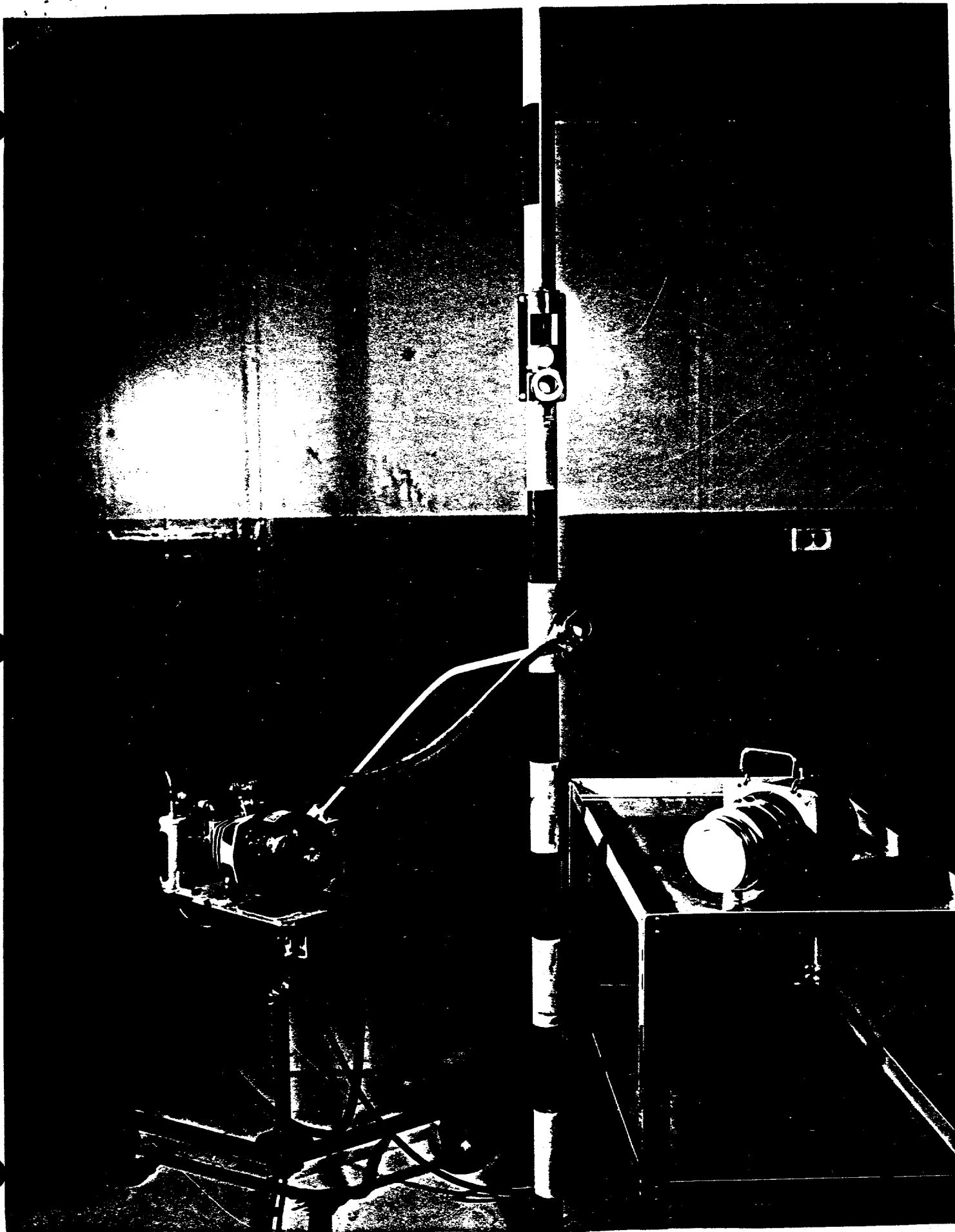


FIGURE 6. AIR SAMPLING EQUIPMENT

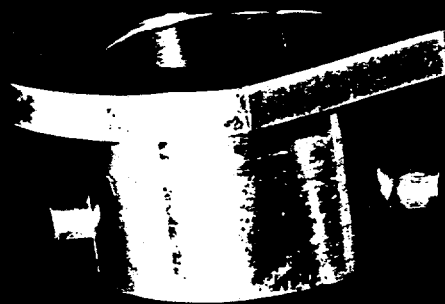


FIGURE 7. EXPLCDED VIEW CF FILTER HOLDER

available outside of research laboratories. Knowing the effective half lives of the radon and thoron daughters, a correction can be applied to the observed counting rates obtained at two separated times but this is, in general, not too accurate.

Another sampling principle is exemplified by the impactor type sampler. This device utilizes the inertia of the particulate matter in the collection process in that an abrupt change in the direction of the air stream causes the heavier particles to impact on a collecting disk. The smaller, and therefore lighter, particles follow the air stream without impaction and, consequently, are not collected. This method has the advantage of passing about 90 per cent of the radon and thoron daughter products since the larger fraction of this activity is carried by the very small inert dust particles. (2) In the cascade impactor, this same principle is utilized to measure particles sizes. The air stream is passed through a series of impactor stages each designed to pass smaller particles than those passed by preceding stages. If the equipment is properly calibrated for particles of given size and type, then one air sampler will collect essentially all of the uranium and separate the particles into the various sizes at the same time. Such a device, consisting of two cascaded stages, is shown in Figure 8.

A variety of portable radiation detectors are available for measuring either radiation fields existing near sources of radiation or radioactive contamination deposited on surfaces. While most of this equipment was originally designed and fabricated in Atomic Energy Commission laboratories, many

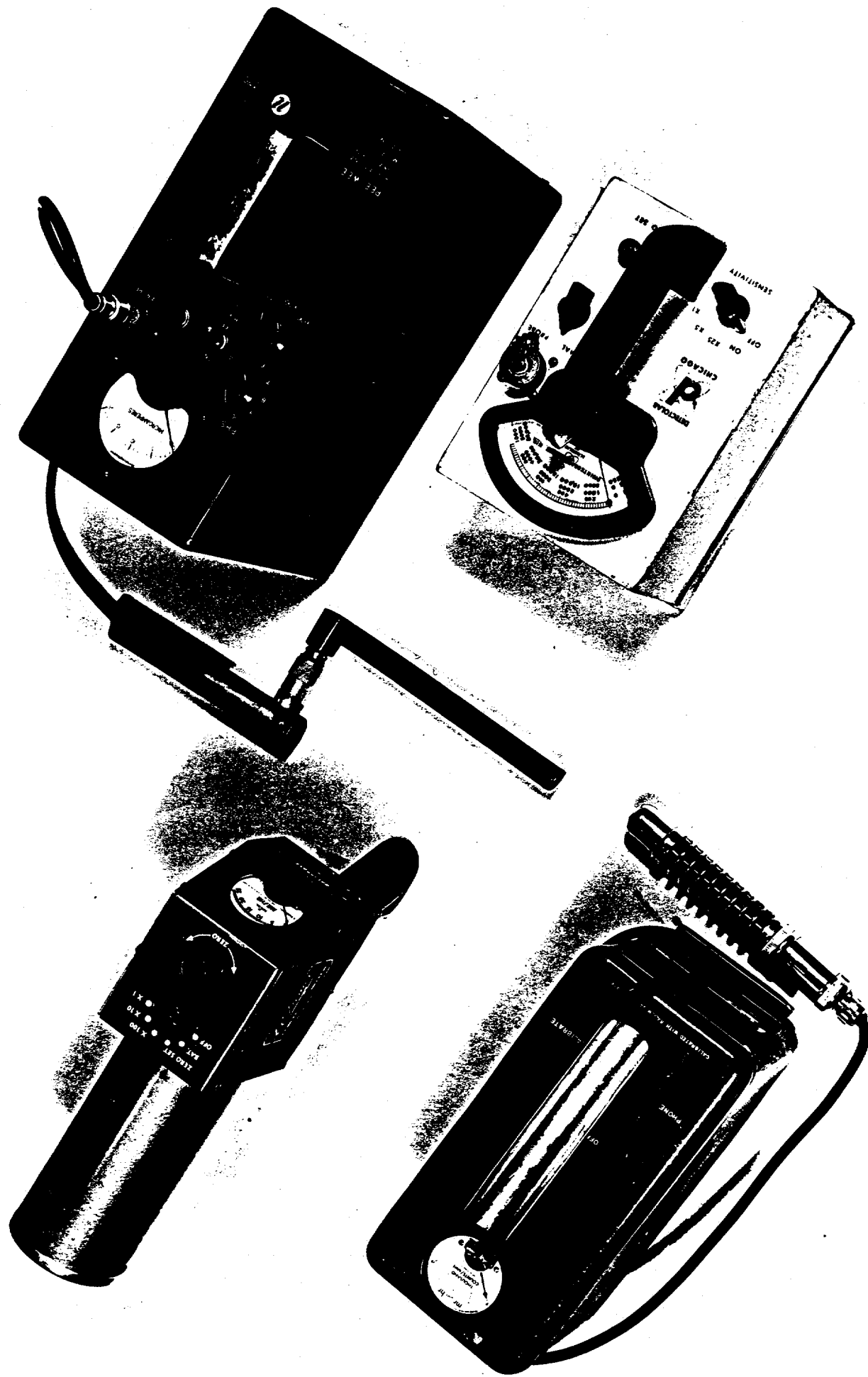


FIGURE 9. PORTABLE RADIATION DETECTORS

are now commercially available. Examples of four typical and different types of such instruments are shown in Figure 9. The ionization chamber instrument in the upper left is used to measure beta and gamma radiation fields at and above the MPL range while the Geiger-Mueller instrument in the lower left measures the same radiations but in the range at and below the MPL. The thin window ion chamber in the lower right is generally used to measure low level surface contamination but has the disadvantage of being unable to distinguish between the various radiations. The proportional counter in the upper right overcomes this disadvantage but at the expense of some bulkiness and added weight. In a typical situation use will be found for all types.

These instruments are calibrated by placing them in varying radiation fields and observing instrument response for a range of known exposures. All are of the rate measuring type in that they measure not the total radiation dose but the rate at which the dose is being delivered. The time integral of the dose rate is the total accumulated dose and this quantity is always measured by an integrating device. Several examples are shown in Figure 10. The film badge and film ring are small devices which can be worn on the body during the period of exposure and later processed to determine the exposure received. Each contains a small piece of radiation sensitive photographic emulsion which darkens upon exposure. The degree of darkening, and thus the total exposure, is determined by measuring the optical density of the developed film. Metal shields are used over part of the emulsion to assist in the determination of the type of exposure and also to decrease the

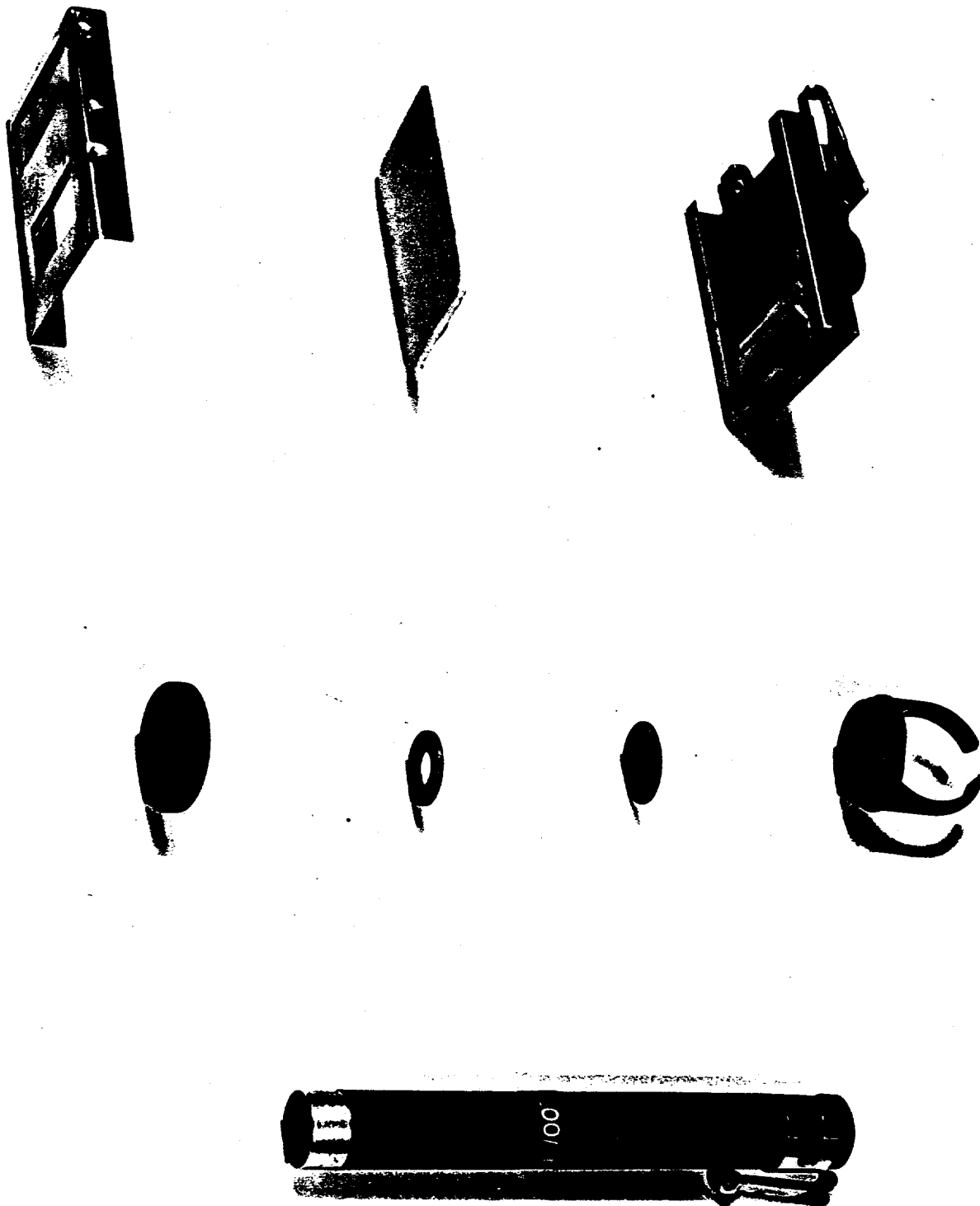


FIGURE 10. INTEGRATING PERSONNEL MONITORS

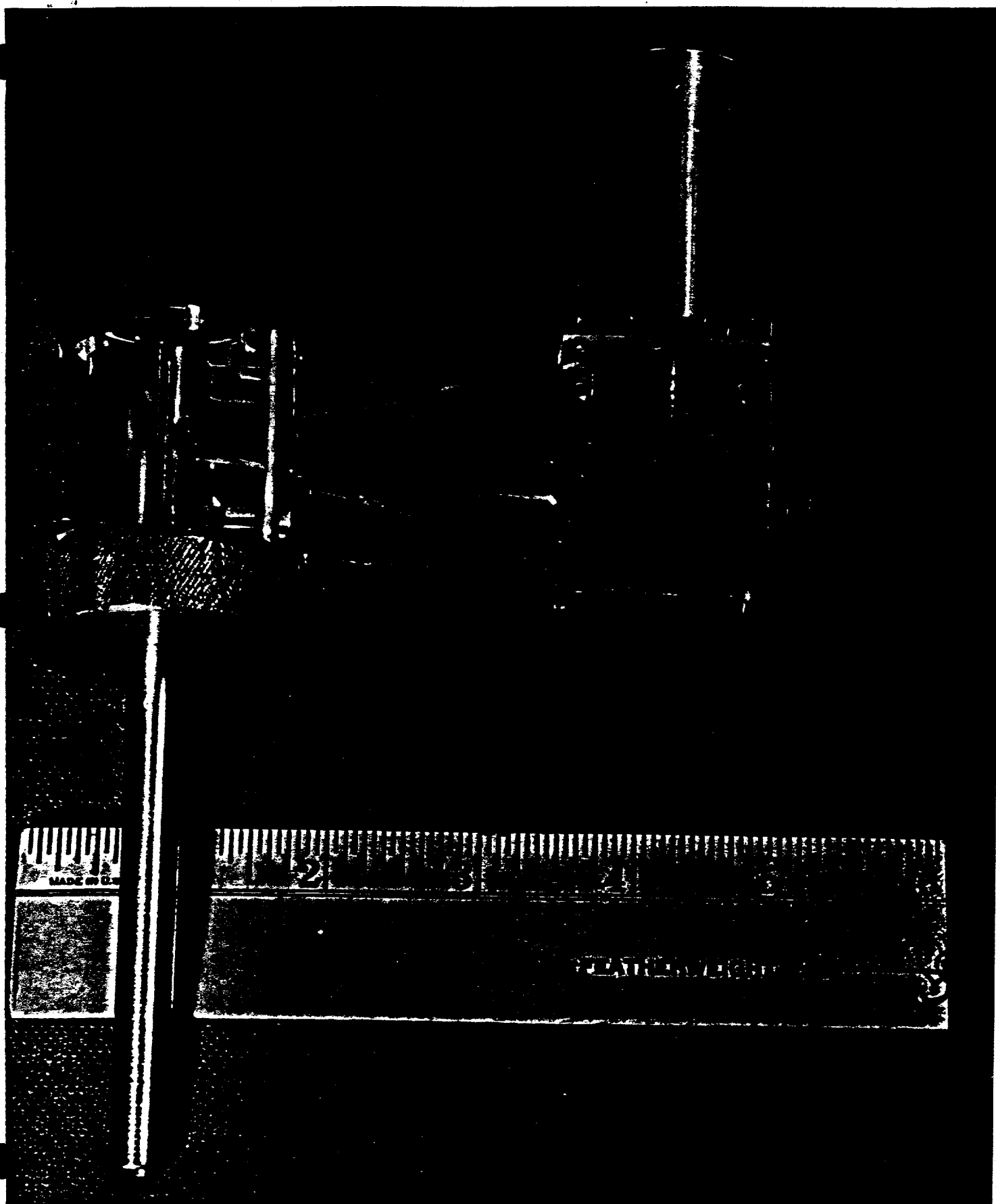


FIGURE 12. TWO-STAGE CASCADE IMPACTOR

energy dependency of the blackening. The pocket chamber is sometimes used as a supplement to the film badge. It has the advantage of not requiring elaborate processing and, thus, can be used over short time intervals, but it possesses the disadvantages of being somewhat delicate and costly.

Personnel monitoring in the case of exposure to alpha radiation cannot be accomplished by the use of film badges. Excretion analysis, in which the individual himself serves as the monitor, affords the only practical approach. With foreknowledge of the distribution and excretion patterns in an average individual, it is possible to relate internal exposure to the amount of uranium found in the urine. This type of monitoring, which required only standard analytical equipment, is a necessity for any program involving the routine handling of uranium or its compounds.

Two analytical methods are in general use. The electroplating method is used for enriched uranium in order to take advantage of the high specific activity. Here the urine is processed to destroy the organic material and then electroplated to deposit the uranium on a metal disk. The disk is then counted in some known efficiency alpha counter to determine the actual amount excreted in the sample. Figures 11 and 12 are two views of a simple electroplating cell used in processing urine samples. A mass arrangement of cells in an installation such as that shown in Figure 13 is capable of handling several hundred samples per week. The alpha counting is usually done with equipment typified by that shown in Figure 14. The one on the left

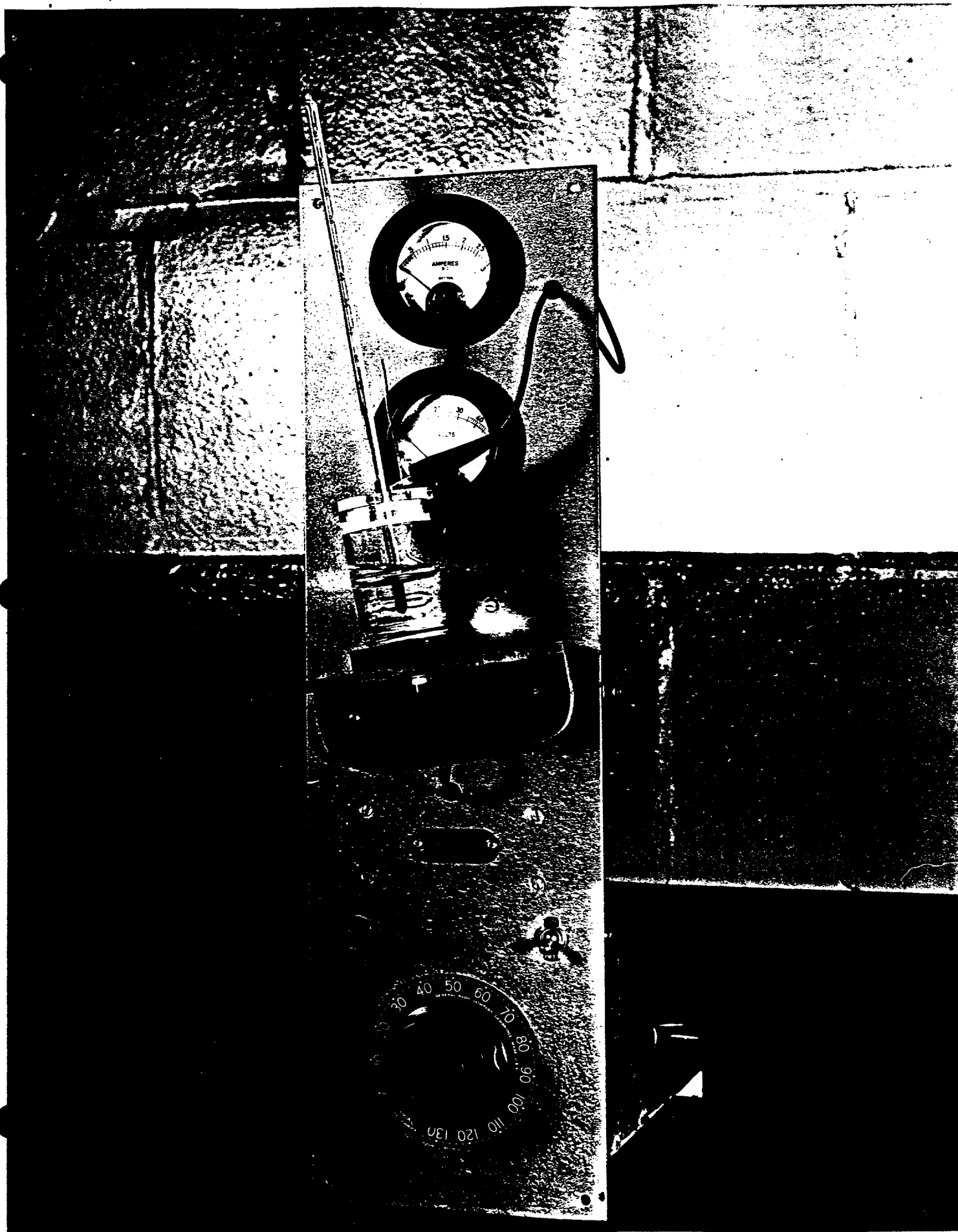


FIGURE 11. ELECTROPLATING CELL

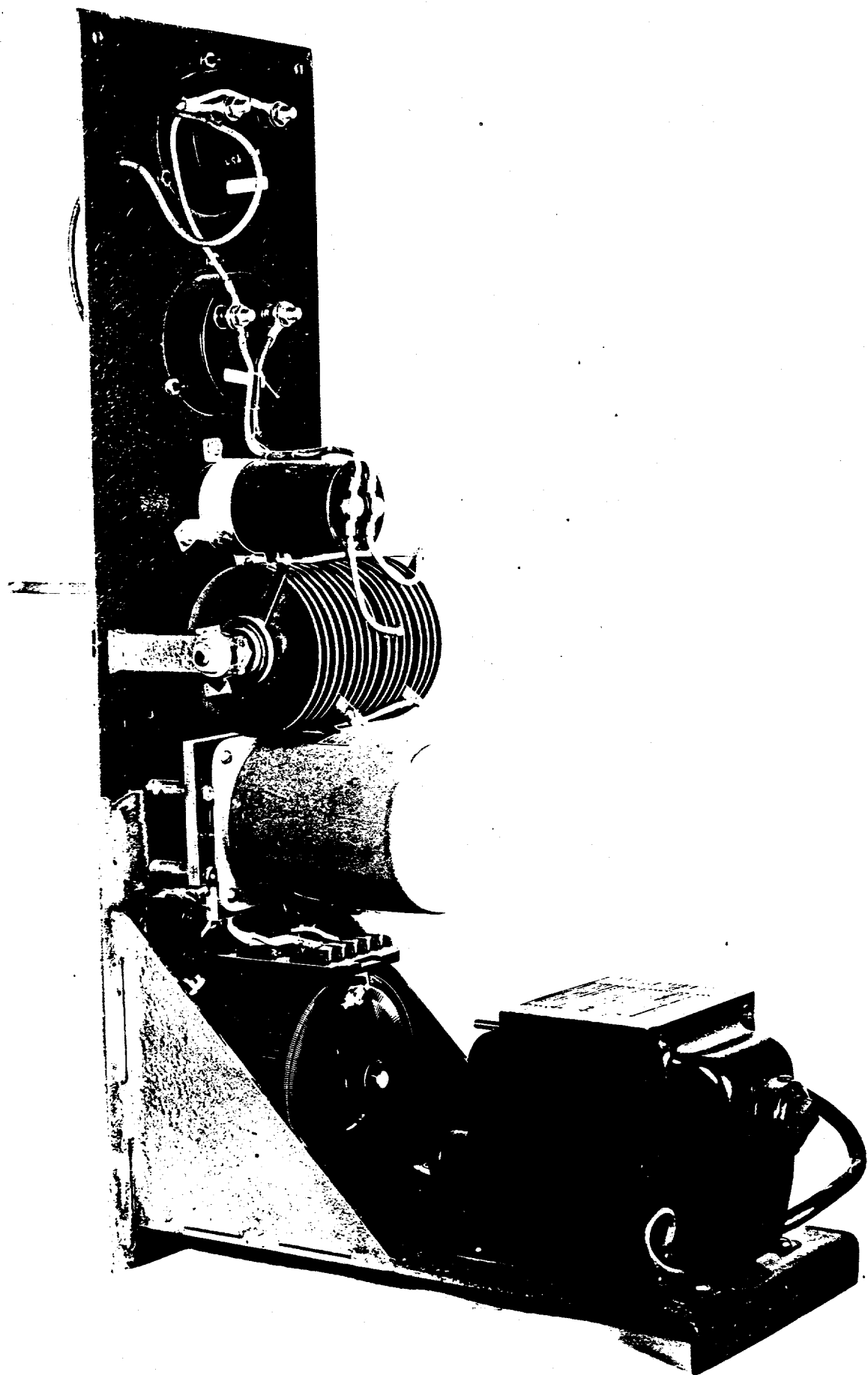


FIGURE 12. REAR VIEW OF ELECTROPLATING CELL

SECTION 12 - TIG AND MAG TUNERS PL ELECTROPLATING CELLS

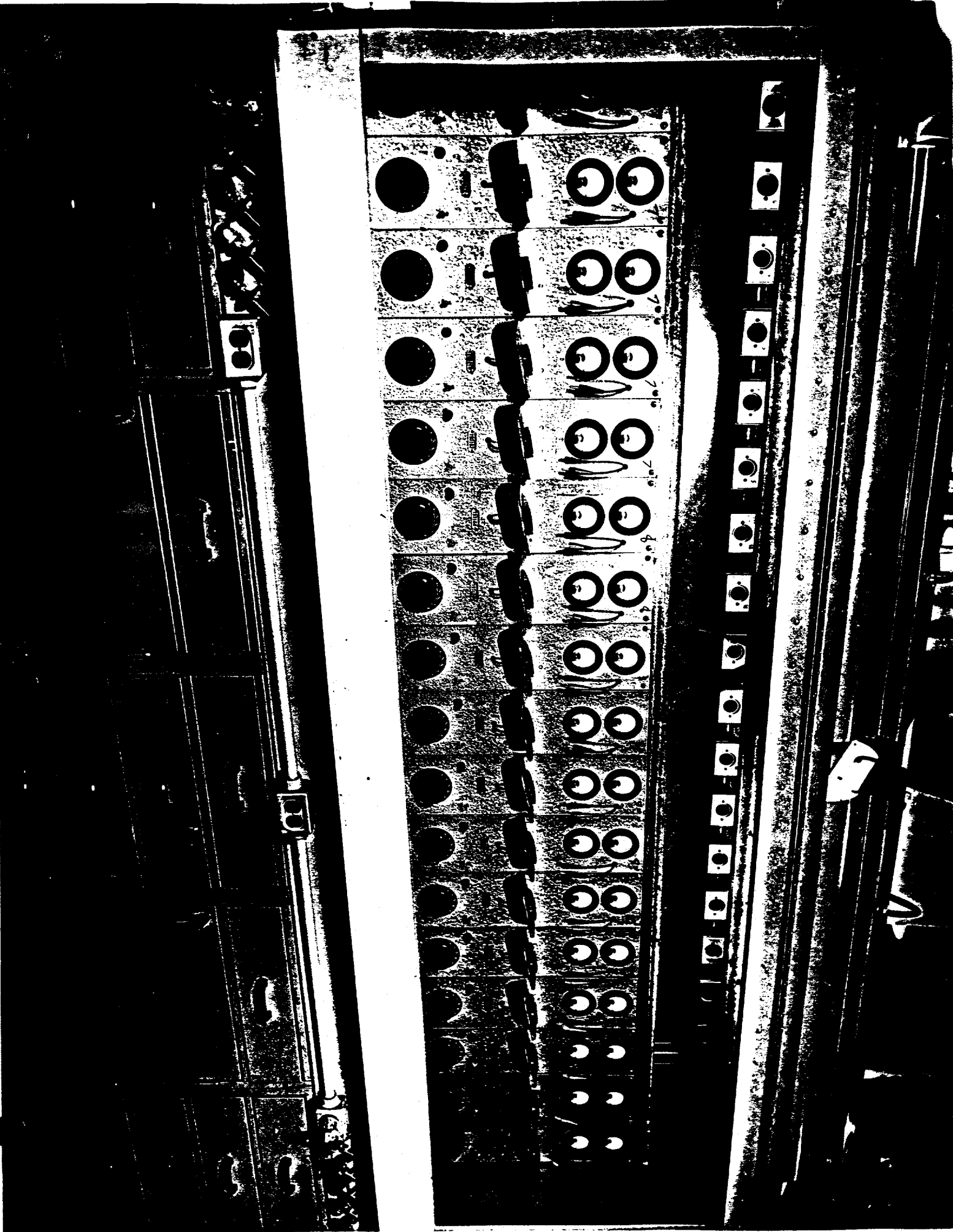
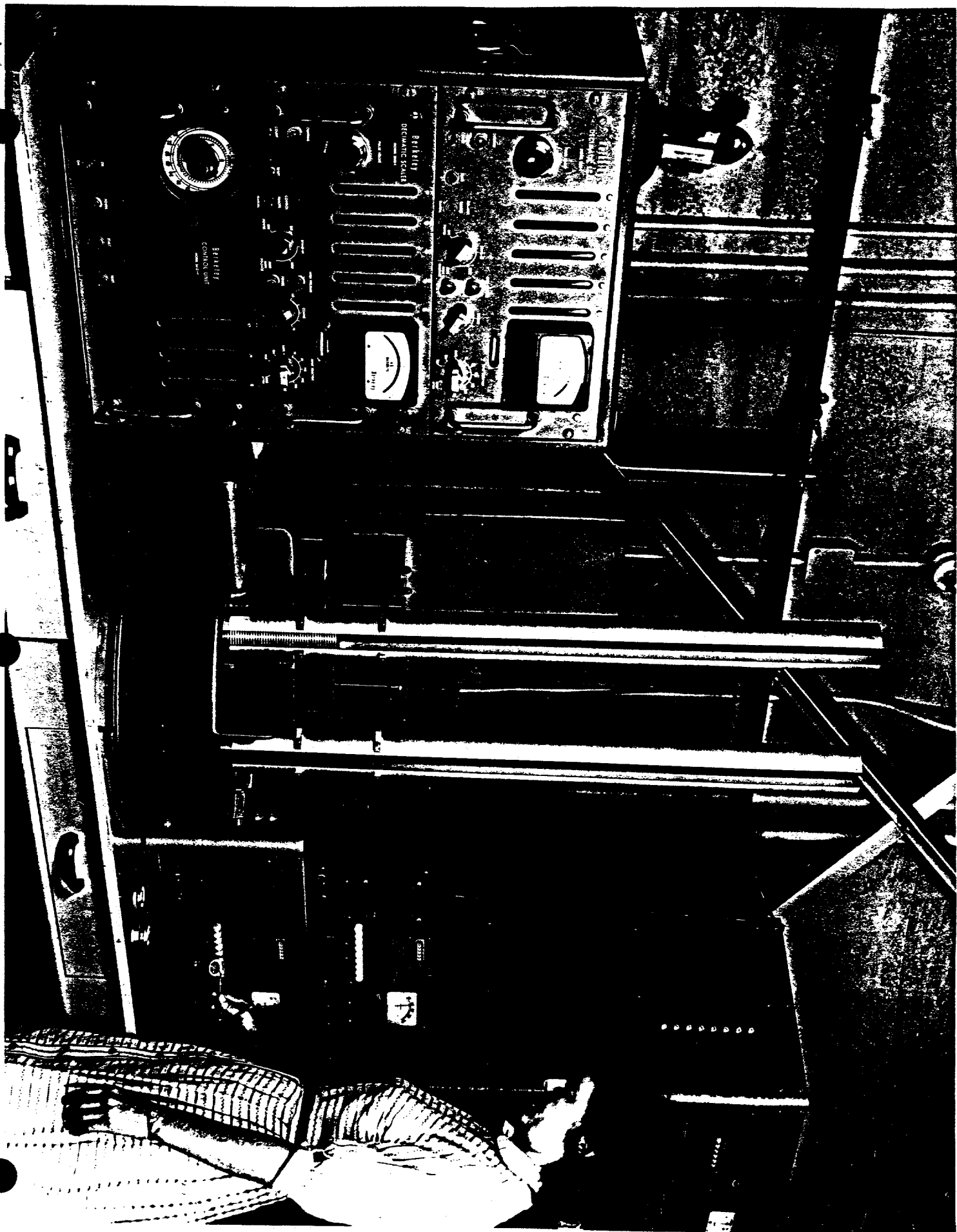


FIGURE 14. ALPHA PARTICLE COUNTING EQUIPMENT



is an automatic scintillation counter capable of analyzing 250 samples in succession without operator attention. The counters on the right use proportional chambers and are perhaps better suited for small programs. For normal uranium samples, in which the specific activity is low, a chemical technique is used. Here the sample is fluxed with a compound to enhance the uranium fluorescence and then exposed to ultra-violet light. The degree of fluorescence is measured using sensitive photoelectric equipment and this quantity then related back to the mass of uranium excreted. A typical installation capable of processing about 500 samples per week is shown in Figure 15.

In the facility using either stack or stream disposal some amount of waste sampling is required. Stack sampling presents special problems but these can be adequately handled by the so-called isokinetic technique. The problem to be overcome is that of distorting the flow pattern and subsequent particle collection by the introduction of a foreign object into the air stream. If the sampling head is designed in such a way that minimum obstruction is presented to the air stream and if the air velocity in the sampler is adjusted to that in the stack, then this distortion is minimized. A typical isokinetic sampling head is shown in Figure 16.

Water sampling presents a special problem in that stream flow is not generally constant and, particularly in the smaller facilities, the waste discharge is not continuous but occurs in discrete batches at varying intervals. This necessitates the installation of continuous sampling equipment. Shown in Figure 17 is such a device which is used in conjunction with a gaging station



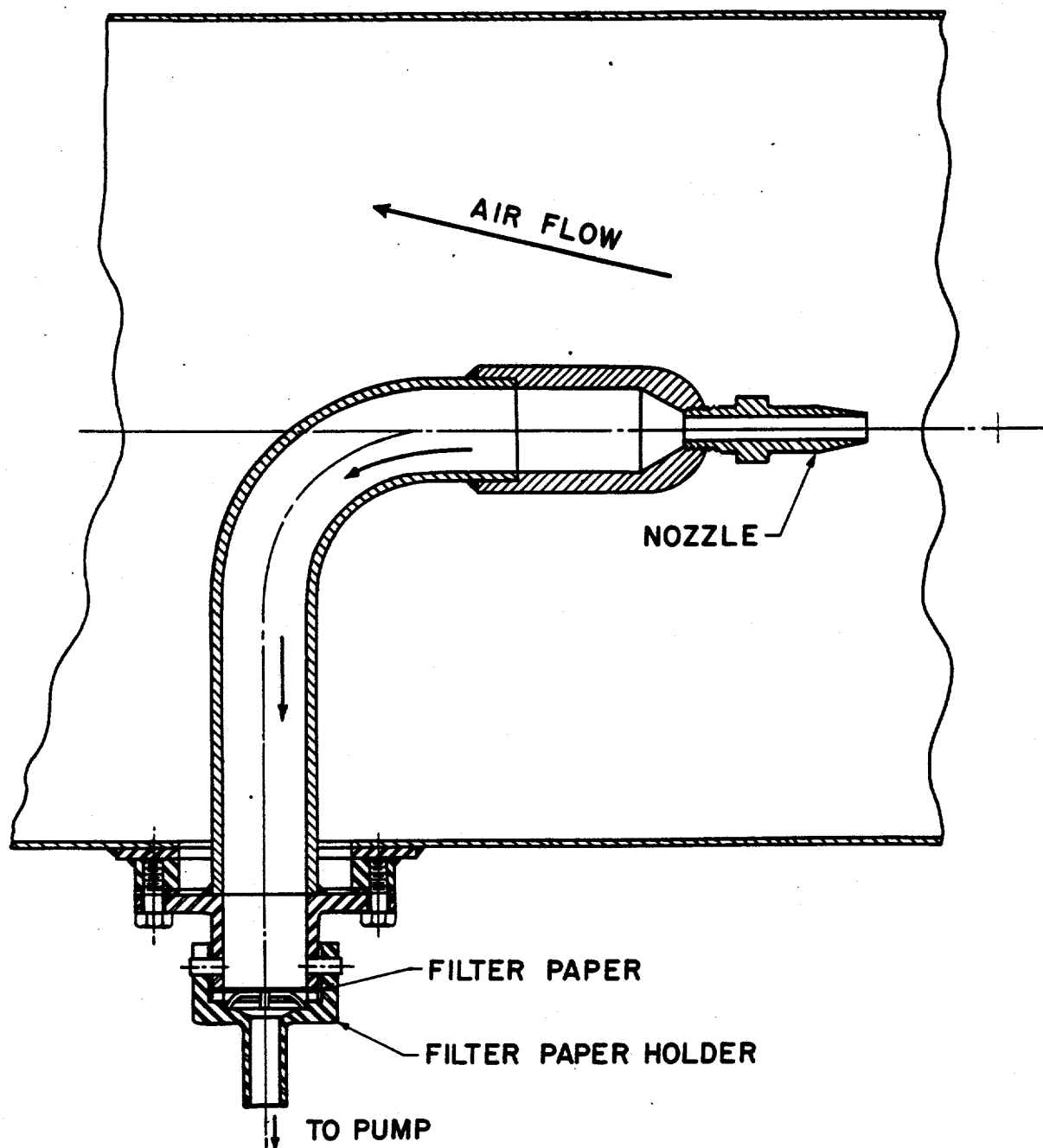


FIGURE 16. ISO-KINETIC SAMPLER ASSEMBLY

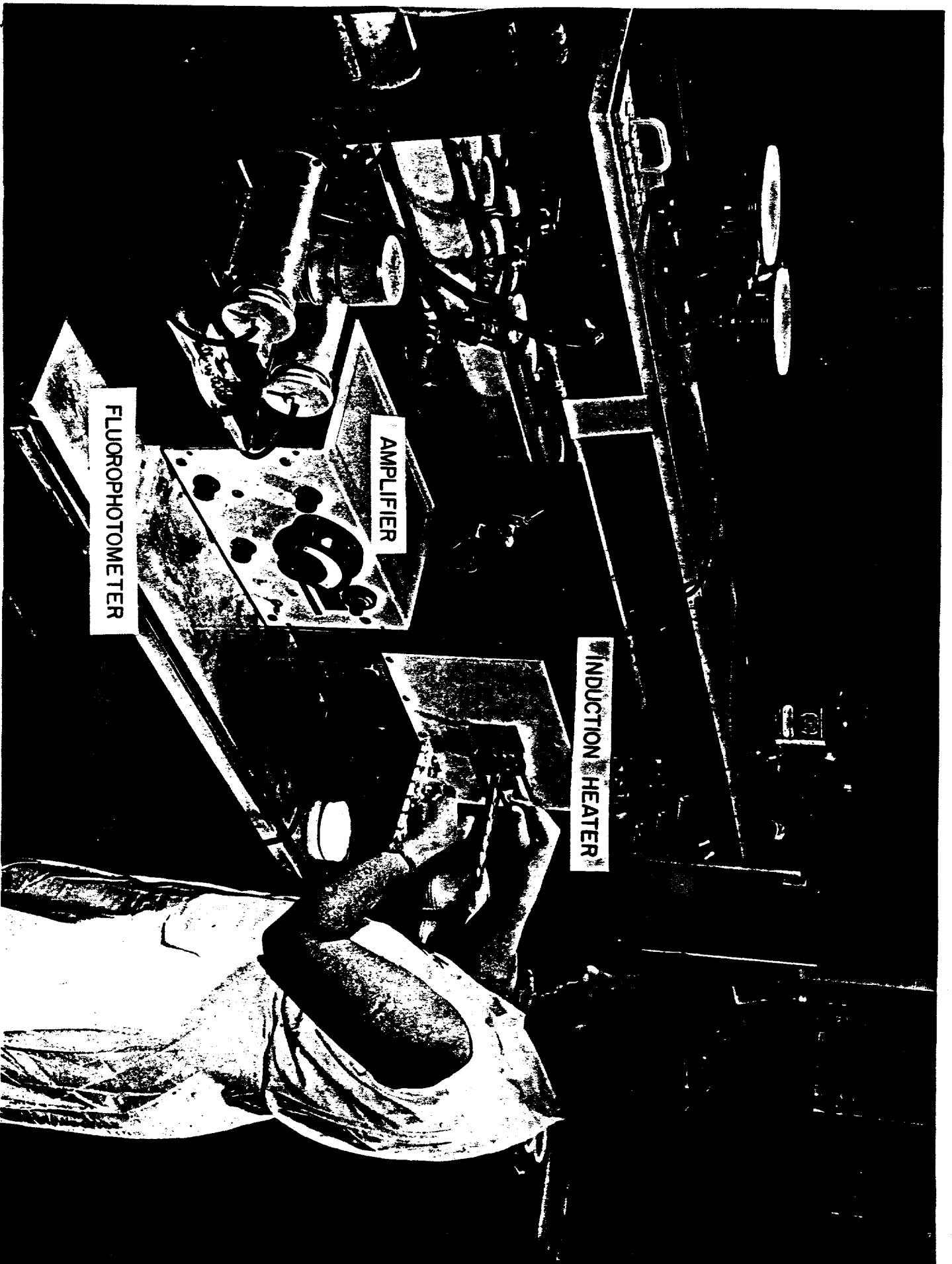
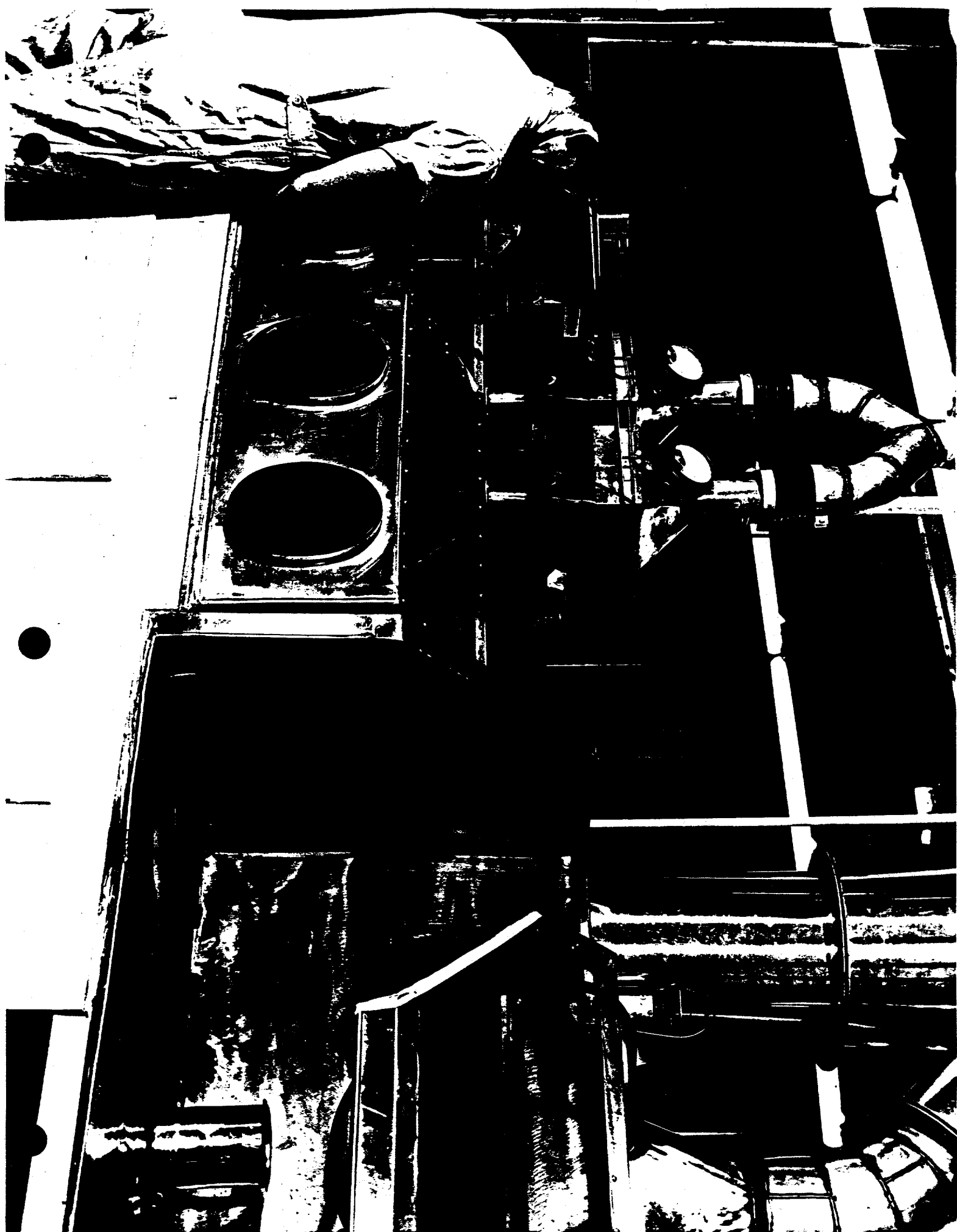


FIGURE 6. MODEL Q FLUOROPHOTOMETER AND ASSOCIATED EQUIPMENT IN USE

to measure stream flow. The sampler can be quite simple; the one shown utilizes a plastic dipper swinging through the water at prescribed time intervals.

Methods and Exposure Control

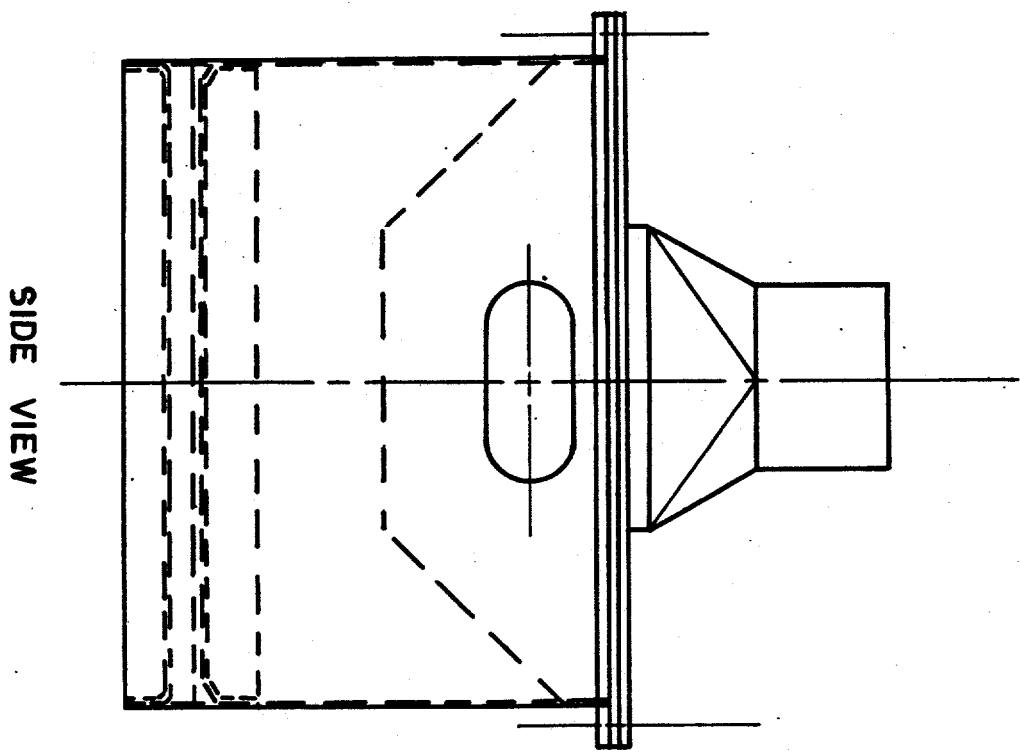
Many of the problems associated with uranium processing can be largely eliminated through proper initial design of the processing equipment. The philosophy is one of containment coupled with proper ventilation control. The ventilation requirements must be tailored to each situation and then rigidly adhered to; rate of air change, direction of flow, and air velocity must all be controlled. In general, systems are designed to carry air from the operator to the work and in such a way that natural settling due to gravity is complemented rather than opposed. Areas of turbulent flow should be avoided where possible. Any unit operations leading to dust generation; such as, grinding, pulverizing and mixing must, in general, be closed or contained. One such container is shown in Figure 18. This dry box is fitted with glove ports to permit flexible operation and also is equipped with a filtered air exhaust. Unless elaborate stack systems are provided, filtering systems must be placed on the exhaust from all confined operations. For enriched materials, this presents no added expense because such systems are usually justified on the basis of material control alone. In any operation, one has recourse to respiratory protective equipment, but, in one sense, the use of such equipment is an admission of engineering defeat.



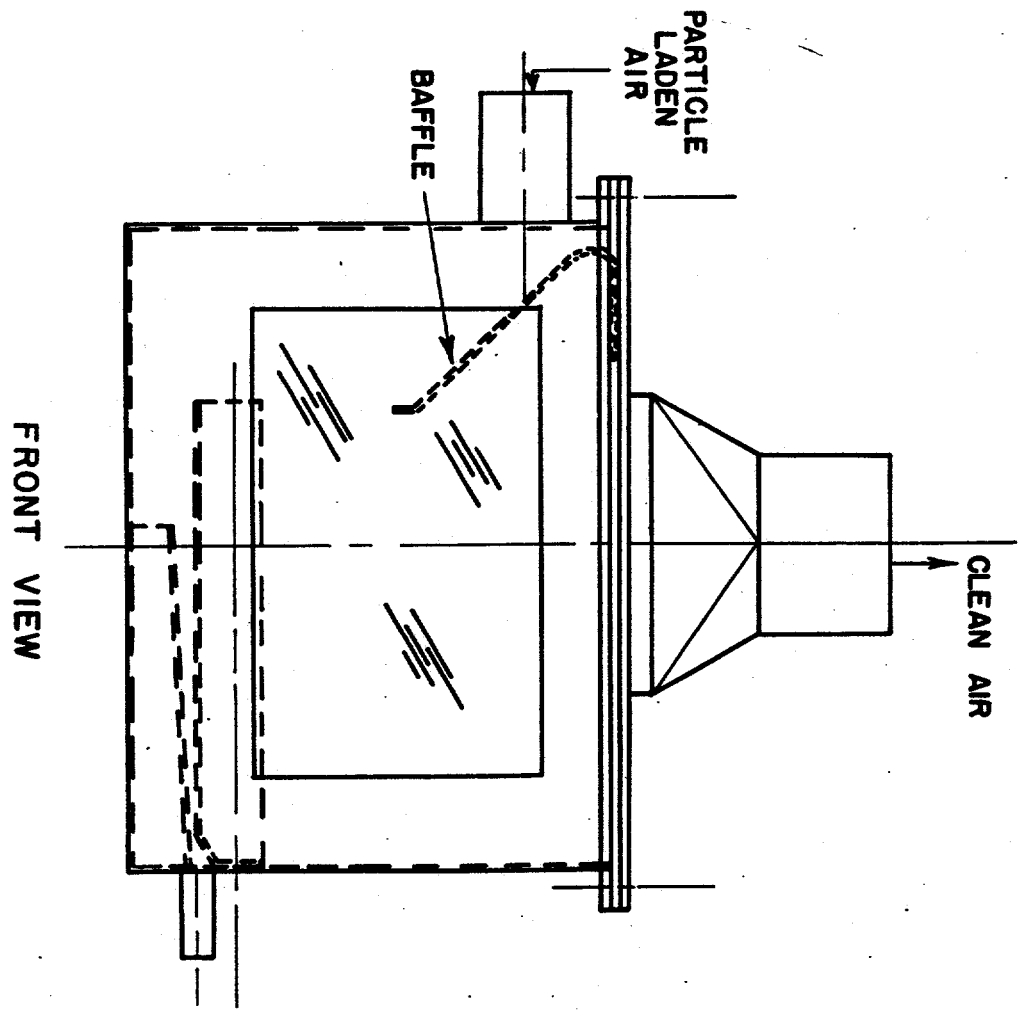
It would be impossible to describe here the proper equipment for each processing step as each case is different. As an example of engineering ingenuity, the particular case of the chip trap may be described. This device, which is shown in Figure 19, is used to remove metal chips from exhaust air streams but the basic principle involved could be applied to any situation involving large particulate matter. The particle laden air stream is introduced into a large settling chamber by means of a deflecting baffle. Since the cross sectional area of the chamber is large compared to that of the influent and effluent air ducts, the air velocity within the chamber is greatly reduced. The combined action of natural settling, which is undisturbed by high air velocities, and the downward initial motion imparted by the baffle serve to remove the larger particles from the air stream.

Proper exposure control through the use of containment and ventilation automatically leads to contamination control. Surface contamination generally is a nuisance and, although hard to evaluate in terms of personnel exposure, it undoubtedly plays some part. Contamination control can be initially attained through the proper design of equipment and facilities but only through good housekeeping can it be maintained. Air contamination is intimately associated with surface contamination and control of one cannot be completely attained without control of the other.

Even with good control over contamination situations will arise in which personnel and equipment become contaminated to a degree requiring action. Fortunately, uranium is quite easily removed; standard detergents are



SIDE VIEW



FRONT VIEW

FIGURE 19. TRAP FOR METAL CHIPS

generally adequate. Wetting agents are of doubtful value, but in severe cases acid washes are sometimes used. Naturally, cases involving the decontamination of personnel should be done only under medical supervision.

In the final analysis, it may be said that the old adage "an ounce of prevention is worth a pound of cure" was never more true than when handling radioactive materials. The prevention of radiation overexposures becomes all the more important when one realizes that, as yet, no medical treatment can prevent the biological damage resulting from severe or continued occurrences. The one most important factor in the successful operation of any processing facility is a constructive attitude, devoid of fear, but founded on a wholesome regard for the problem.

BIBLIOGRAPHY

1. Neuman, W. F., "Urinary Uranium as a Measure of Exposure Hazard", The University of Rochester, UR-82, (1949)
2. Tait, G. W. C., "Determining Concentration of Airborne Plutonium Dust", Nucleonics, 14, 1, 53 (1956)

Selected References for Basic Information:

1. Parker, H. M., "Health Physics, Instrumentation, and Radiation Protection", United States Atomic Energy Commission Report MDCC-783, (1947)
2. Cantril, S. T. and Parker, H. M., "The Tolerance Dose", United States Atomic Energy Commission Report MDCC-1100, (1947)
3. Evans, R. D., "Health Physics: Instrumentation and Hazard Evaluation", The Science and Engineering of Nuclear Power, Chapter 16, (1947)
4. Taylor, L. S., "The Basis for Standards for Radiation Protection", Journal of the Society for Nondestructive Testing, 14, 1, 10-15, (1956)
5. National Committee on Radiation Protection, "Safe Handling of Radioactive Isotopes", National Bureau of Standards Handbook 42, (1949)
6. International Commission on Radiological Protection, "Recommendations of the International Commission on Radiological Protection and of the

International Commission on Radiological Units 1950", National Bureau of Standards Handbook 47, (1951)

7. National Committee on Radiation Protection, "Control and Removal of Radioactive Contamination in Laboratories", National Bureau of Standards Handbook 48, (1951)
8. National Committee on Radiation Protection, "Radiological Monitoring Methods and Instruments", National Bureau of Standards Handbook 51, (1952)
9. National Committee on Radiation Protection, "Maximum Permissible Amounts of Radioisotopes in the Human Body and Maximum Permissible Concentrations in Air and Water", National Bureau of Standards Handbook 52, (1953)
10. Ehrlich, Margarete, "Photographic Dosimetry of X- and Gamma Rays", National Bureau of Standards Handbook 57, (1954)
11. National Committee on Radiation Protection, "Permissible Dose from External Sources of Ionizing Radiation", National Bureau of Standards Handbook 59, (1954)
12. British Institute of Radiology, London, 1955, "Recommendations of the International Commission on Radiological Protection (Revised December 1, 1954)", British Journal of Radiology Supplement Number 6, (1955)